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SYSTEMS AND COSTS TO CONTROL HYDROCARBON EMISSIONS FROM STATIONARY SOURCES

ENVIRONMENTAL PROTECTION AGENCY

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Costs associated with techniques for controlling hydrocarbon emissions from certain important stationary sources are discussed. Sources discussed include organic solvent usage, petroleum refining and marketing, and certain petrochemical manufacturing operations.

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SYSTEMS AND COSTS TO CONTROL HYDROCARBON EMISSIONS FROM STATIONARY SOURCES

EMISSION STANDARDS AND ENGINEERING DIVISION

U.S. ENVIRONMENTAL PROTECTION AGENCY
Office of Air and Waste Management
Office of Air Quality Planning and Standards
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CONTENTS

	Page
Summary	1
Introduction	3
Organic Solvent Usage	
Degreasing	5
Dry Cleaning	9
Graphic Arts	13
Surface Coatings	17
Petroleum Refining and Marketing	
Petroleum Refineries	21
Petroleum Product Liquid Storage	23
Gasoline Distribution	26
Petrochemical Manufacture	
Carbon Black	32
Acrylonitrile	34
Formaldehyde	36
Ethylene Dichloride	39

TABLES OF EMISSIONS AND ECONOMICS

	Page
Table 1 - Hydrocarbon Emissions from Stationary Sources	3
Table 2 - Degreaser Control Costs	7
Table 3 - Solvent Emissions from Typical Dry Cleaning Plant	10
Table 4 - Estimated Emissions from Various Types of Printing Plants	15
Table 5 - Emissions Control Costs in the Graphic Arts Industry	16
Table 6 - Investments - Floating Roof vs. Fixed Roof Tanks	24
Table 7 - Control Costs for Retrofiting Fixed Roof Tanks to Covered Floating Roofs	25
Table 8 - Hydrocarbon Emissions from an Uncontrolled Gasoline Distribution System	27
Table 9 - Control Costs per Service Station Dispensing 25,000 Gal/month of Gasoline	29
Table 10 - Estimated Emissions from a 200 Million Pounds per Year Acrylonitrile Plant	34
Table 11 - Emissions from an Uncontrolled 100 Million Pounds Per Year Formaldehyde Plant	37
Table 12 - Emissions from an Uncontrolled 700 Million Pounds Per Year Ethylene Dichloride Plant	39

SUMMARY

Organic solvent usage has been identified as a major source of hydrocarbon emissions. However, the technical information necessary to evaluate many of the emission control systems that could be employed in this area, or their emission control costs, is marginal or completely lacking in most cases. For example, it is evident that the adoption of water-based coatings would significantly reduce emissions from industrial surface coatings operations. However, neither the extent to which water-based coatings could be used in place of organic solvent-based coatings, nor the costs associated with converting large surface coating operations to the use of these coatings, is known. Furthermore, the time tables necessary for industrial conversion are not known, nor is the impact on small job operators known. The same is true in the graphic arts industry. While adoption of a regulation patterned after the LAC-APCD Rule 66, will accelerate development of reformulated surface coatings and emission control technques for smaller operations such as degreasing, an assessment at this time of the impact of such a regulation on the national level is not possible. Consequently, various alternative regulations, more stringent than Rule 66, will be quite difficult to justify.

Another area of concern is gasoline distribution and marketing. In this area, while most of the technology is well demonstrated and generally available, some is still under development. Consequently, control of this source should be implemented in two phases. Phase I would reduce hydrocarbon emissions by about 55 percent, by controlling emissions during gasoline loading into tank trucks and trailers and during gasoline deliveries to service stations. Although a number of problems such as demonstration of safe wintertime operation of vapor recovery units and adaptability and scale down of these units to small loading racks remain to be resolved, this should not delay rapid implementation of this phase.

Phase II, control of emissions during the refueling of automobiles, would reduce hydrocarbon emissions by an additional 30 to 35 percent, but is far more complex than Phase I. Many basic technical problems such as successful design of a tight-fit gasoline dispensing nozzle, standardization of automobile fuel tank fill pipes, and establishing the reliability and safety of small vapor recovery units at gasoline service stations remain to be resolved.

INTRODUCTION

Table 1 summarizes the major stationary sources of hydrocarbon emissions which have been identified.

Table 1

Hydrocarbon Emissions from Stationary Sources[1]

Source	Range of Emissions
Organic Solvents Usage	(10^6 tons/year)
Degreasing	0.6 - 0.9
Dry Cleaning	0.3 - 0.4
Graphic Arts	0.2 - 0.3
Surface Coatings	3.0 - 4.0
Petroleum Refining and Marketing	3.6 - 4.0
Chemical Industries	1.0 - 1.4

Industrial surface coating operations and architectural coating applications, are estimated to contribute equally to the emissions from surface coatings shown in Table 1. Petroleum refining and marketing, which represents one of the largest emission sources, includes three subsource categories. These are petroleum refining (excluding petroleum storage), petroleum storage and gasoline distribution and marketing.

A major study is currently in progress to identify emissions and available control techniques within the petrochemical industry. Screening studies of 40 petrochemical processes indicate that 30 of these processes emit about 600,000 tons per year of hydrocarbons. In-depth studies have been completed on processes for the manufacture of carbon black, acrylonitrile, formaldehyde and ethylene

dichloride. The manufacture of these four petrochemicals accounts for about 40 percent of the 600,000 tons per year of hydrocarbon emissions which have been identified. The emission control systems to limit these emissions and the costs associated with these systems are reviewed in this report.

DEGREASING

Degreasing is a term broadly applied to the use of various organic solvents to dissolve and remove grease and soils from metal objects. If the solvent is near or at room temperature, the process is termed "cold solvent cleaning"; if the solvent is maintained at its boiling point, the process is referred to as "vapor degreasing". Vapor degreasing accounts for most of the solvent emissions from this source category.

Most vapor degreasers consist of an open tank equipped with a liquid heater, a vapor zone above the liquid surface, and a narrow water-cooled jacket (chiller) near the top of the tank to restrict the vapor zone. A free board area extends beyond the vapor zone and this also restricts vapor emissions to some extent. A hood is normally installed above the degreaser to collect and discharge the solvent vapors which escape to the atmosphere.

When cool metal objects are lowered into a vapor degreaser, solvent vapors condense on the metal surfaces until the metal reaches the vapor temperature. Soils such as dirt, metal cutting fines and grease are washed off the metal objects by the condensing vapors and drop back into the tank.

Emissions

The vapor escape rate from an open top degreaser is estimated to be in the range of 0.5 to 1.0 lb/hr-ft^2 of open tank area.[2] Vapor emissions are accelerated by air currents passing over the open or partially open degreaser, by opening the lid if the degreaser has one, by spraying the articles with solvent, and moving the articles in and out of the degreaser. However, there is little data available on various types of degreasers to estimate typical hydrocarbon emission factors.

Control Systems and Costs

Most emission control techniques for degreasers attempt to prevent hydrocarbon emissions from the tank through improved operating techniques, rather than control the exhaust gases. This includes instrumentation to prevent excessive overboil in the degreaser by temperature controllers and interlock systems to prevent operation of the unit if there is not adequate coolant flow through the cooler jacket. These techniques have been cited as capable of reducing emissions by about 20 to 30 percent.[3] The use of a refrigerated cooling jacket is cited as capable of reducing emissions by about 10 percent and if tank lids are installed, it is estimated that emissions can be reduced by about 50 percent.[3]

The solvents primarily used within the industry are trichloroethylene, perchloroethylene and 1, 1, 1 trichloroethane. The industry prefers trichloroethylene in lieu of 1, 1, 1 trichloroethane. In the presence of moisture, 1, 1, 1 trichloroethane can hydrolyze to hydrochloric acid which can harm the products being degreased.

Emission control costs are summarized in Table 2 for three alternatives. These costs are based on an open tank vapor degreaser operating 40 hours per week with uncontrolled emissions of 660 pounds per week of trichloroethylene (16.5 lbs/hr or 0.825 lbs/hr-ft^2). In Case 1, the degreaser is fitted with temperature control instrumentation and operated according to

procedures recommended by the solvent manufacturer. This reduces solvent emissions to 0.5 lb/hr-ft^2 and results in a net annual savings. In Case 2, the degreaser is fitted with temperature control instrumentation and refrigerated cooling coils around the top of the vessel. This case also shows a net annual savings due to the value of the recovered solvent. Case 3 is based on the best practical control, carbon adsorption, which achieves a 90 percent reduction in emissions. Although many existing degreasers could probably not be adequately vented to capture all of the evaporated solvent, Case 3 assumes that no ventilation problems exist. It should be noted however, that not many plants would find it economically feasible to make the large investment necessary for carbon adsorption (relative to the basic investment necessary for the degreaser), if other alternatives to degreasing were available.

Table 2

Degreaser Control Cost

Control Equipment	Case 1[3,4] Temperature Controls plus Good Operation	Case 2[3,4] Case 1 plus Refrigerated Coils	Case 3[5] Carbon Adsorption
Emissions to Atmosphere lb/hr-ft^2	0.50	0.41	0.08
Capital Cost[a] ($)	300	3000	19,500
Net Annual Cost or (Savings) ($/yr)	(1,428)	(1,070)	620

a The basic degreaser unit cost is about $2,000.

Rules and Regulations

In some State Implementation Plans (SIP's) proposed by the States and EPA, degreasing operations have been isolated from other organic solvents usage. A number of SIPs permit only perchloroethylene or saturated halogenated hydrocarbons to be used in degreasing. This bans the use of trichloroethylene, regardless of the control efficiencies that could be achieved.

DRY CLEANING

Most industrial dry cleaners use either petroleum solvents, which are referred to as "Stoddard" solvents or, perchloroethylene, which is referred to as "Perc". Stoddard solvents are flammable mixtures of paraffins, cycloparaffins, and aromatics, while Perc is a non-flammable chemical. Solvent consumption is not indicative of the tonnages of fabrics dry-cleaned by each solvent. Since Stoddard solvent has a value of $0.25 per gallon and Perc has a value of $1.39 per gallon, economic incentives have prompted the development of systems for the recovery of Perc, but not for Stoddard solvents.

Emissions inventory data on dry cleaning operations, with a reasonable breakdown of fabric tonnages cleaned by each solvent, and the size distribution of dry cleaning establishments is lacking. Preliminary estimates indicate a 55/45 split between Perc and Stoddard usage in terms of the fabric tonnages cleaned, but this has not been validated. It is doubtful however, if many new plants using Stoddard solvents will be constructed in the future, since most Stoddard solvents have a minimum flash point of 100°F. For safety reasons, a number of local zoning commissions and insurance underwriters are prohibiting the use of Stoddard solvents.

In a typical dry cleaning plant using Stoddard solvents, the washer and drier are usually separate pieces of equipment. The solvent extracted by centrifuging in the washer is filtered and reclaimed by vacuum distillation. The muck remaining, which contains unrecovered solvent, is disposed of by whatever means are available. The solvent remaining in the clothing is sent to a drier, evaporated and exhausted to the atmosphere.

Dry cleaning plants utilizing perchloroethylene normally unitize the cycle by combining the washing and drying in one piece of equipment. While this may reduce dry cleaning thruput per unit, a greater control over perchloroethylene consumption is possible. Perchloroethylene extracted after washing is filtered and recovered by distillation. In addition, cookers are normally operated to recover solvent from the filter muck.

Emissions

Solvent emissions from a typical uncontrolled industrial dry cleaning plant with a 50 pound washer handling 500 pounds of fabric per day are shown in Table 3. The Stoddard plant is a washer-extractor (cold transfer) type equipped with a regenerative filter and the Perc plant is a conventional utilized system (dry to dry) equipped with a distillation still, regenerative filter, cooker and condenser.

Table 3

Solvent Emissions From Typical Dry Cleaning Plants[6]

Solvent	Stoddard	Perchloroethylene
Emissions to Atmosphere, lbs/day		
Evaporated at Tumbler	95	14
Other Loses, lbs/day		
Retained in Filter Muck	25	22
Retained in Still Residue	5	8
Total Loses, lbs/day	125	44

Control Systems and Costs

The value of Perc makes its recovery economically attractive. A number of manufacturers offer packaged carbon adsorption systems, which permit recovery of essentially all the Perc not recovered by condensers on the distillation still and muck cooker.

A carbon adsorption system for the Perc dry cleaning plant presented in Table 3 would cost about $1,400.[7] Perc emissions would be reduced by about 98 percent and the value of the recovered solvent would offset the annual costs for this equipment.[7] Thus, installation and operations of the adsorber would impose no increased costs on the dry cleaning plant.

Control of solvent emissions from a dry cleaning plant using Stoddard solvent is not as straightforward. Carbon adsorption, which could probably reduce emissions by about 90 percent, appears to be the logical emission control technique. However, the low value of Stoddard solvent makes the economics of recovery unattractive except for large plants. In addition, the physical properties of Stoddard solvents make them more difficult to adsorb and desorb than Perc. Consequently, carbon adsorption systems have not been applied to small Stoddard dry cleaning plants of the size shown in Table 3. For a Stoddard dry cleaning plant about four times the size of the plant shown in Table 3, one manufacturer has estimated the capital costs for a carbon absorption system to be about $17,000.[8] After taking credit for the recovered solvent, the annual emission control costs, based on a one shift operation, would be about $2,100 or approximately $0.22 per pound of Stoddard solvent recovered.[8]

Substitution of perchloroethylene for Stoddard solvent may be impractical at many existing plants because of equipment differences. Perchloroethylene

is corrosive under various conditions and cannot be handled in Stoddard washers and driers which are designed for non-corrosive solvents. This means that essentially all of the equipment in a conventional Stoddard dry cleaning plant might have to be replaced if Perc were substituted. Preliminary estimates of this cost are about $30,000 per plant.[8] Furthermore, the dry cleaning associations have indicated that perchloroethylene cannot be used to dry clean all fabrics.

Rules and Regulations

Rule 66 of the LAC-APCD exempts perchloroethylene from control and allows the use of Stoddard solvent without control if its aromatics content is less than 8 percent and emissions do not exceed 3000 lbs. per day. Variations of this rule appear in many State Implementation Plans and recent EPA proposals. However, this approach of exempting perchloroethylene and Stoddard solvents from control, has come under question by the Chemistry and Physics Laboratory (ORM, NERC, Research Triangle Park, N. C.). Recent data gathered from smog chamber experiments indicates that both Stoddard solvents and perchloroethylene are photochemically "reactive."

GRAPHIC ARTS

Commercial printing within the graphic arts industry refers to the following printing processes: lithography; letterpress; gravure; flexography and screen. The solvents used in the various ink formulations are a source of hydrocarbon emissions. Heat set inks, such as those used in lithography and letterpress printing, generally contain 30 to 40 percent solvent. Gravure, flexographic and screen inks however, generally contain 55 to 75 percent solvent.

Lithography is the fastest growing segment of the commercial printing industry. The lithographic process transfers ink from an image plate to a rubber surface, which in turn transfers the ink to the surface being printed. The non-image area on the image plate is essentially at the same level as the image area. However, the non-image area is wet by water only, while the image area is wet by ink. Lithography is widely used for printing newspapers, magazines, and books. In addition, most all metal decorating operations have adopted lithography.

In the letterpress printing process, the ink is transferred directly to the material being printed by the image plate. The image area of the plate is raised relative to the non-image area and only the image area is wet with ink. Letterpress printing was the major printing process employed within the commercial printing industry until recently. In the last few years, the use of letter press printing has declined, while the use of lithography has increased. Letterpress printing is still widely used however, to print newspapers, magazines and periodicals.

Gravure printing is widely used to print newspapers and advertising

supplements and catalogs, and various packaging materials such as gift wrap and labels. In this type of printing, the image area on the image plate is recessed relative to the non-image area. As ink is transferred to the image plate, it is picked up by a "doctor blade". The image plate is then used to transfer ink directly to the surface of the material being printed.

Flexography is essentially a letterpress process employing rubber image plates. Although the flexographic process is widely used to print packaging materials, its major use is in the printing of paper-back "pocket" books. Most all paper-back "pocket" books are now printed by flexography.

Screen printing represents one of the smallest segments of the printing industry. It is however, employed extensively in the specialized area of printing paper and cardboard signs, posters, and displays. Although not normally considered as part of the commercial printing industry, a rapidly growing application of screen printing is in the production of microcircuits. The image area in screen printing, is a fine screen through which ink can be forced. Non-image areas are produced by coating or impregnating the screen with a wax to prevent the flow of ink through that portion of the screen.

Emissions

Emissions vary considerably from one printing job to the next and are dependent on the solvent content of the ink used, press operating speed, and ink coverage. Presses do not normally operate continuously and may be idle for considerable periods of time during plate changing, press repair, and slack periods of work. Although emissions are difficult to estimate, a range of emissions is presented in Table 4 for the various types of printing operations.

Table 4

Estimated Emissions from Various Types of Printing Plants[9,10]

Process	lbs/hr	1970 Percent of Market Sales
Gravure (Roto)	50 - 250	6.6
Flexography (heatset inks)	2 - 15	4.3
Lithography		
Web offset	4 - 25	51.0
Metal decorating	4 - 5	---
Letterpress (heatset inks)	2 - 15	35.5
Screen printing	low	2.6

Control Systems and Costs

Incineration is the most commonly employed control technique for reducing emissions from commercial printing operations. Although incineration can be applied to all printing processes, it is more adaptable to those processes which use ovens to evaporate the solvent from the ink. If the inks used in the process are free from materials which could poison or form a coating on the various catalysts employed, both catalytic and thermal incineration is effective.

Adsorption can be employed as an emission control technique to some extent. Normally, adsorption is practical only where solvent emissions have not been partially oxidized. Consequently, it is generally not adaptable to driers or ovens. Most applications of adsorption have been in gravure printing, where large amounts of solvent are emitted.

Emission control costs for reducing ink solvent emissions from lithographic and letterpress printing operations, based on incineration of the drying oven off-gases, are summarized in Table 5. Although these costs are considered typical, the wide range of conditions which exist within the commercial printing industry could result in substantial deviations from these costs for specific installations. The costs for the web offset lithographic process and the letterpress process, reflect incineration of the emissions from one drying oven. The costs for the metal decorating lithographic process, reflect incineration of the emissions from two ovens.

Table 5

Emissions Control Costs in the Graphic Arts Industry[11]

Process	Web Offset Lithography/ Letterpress		Metal Decorating Lithography	
Emissions (lbs of carbon/hr)	5		40	
Gas volume (scfm)	5,000		10,000	
Control Device	Thermal Incinerator	Catalytic Incinerator	Thermal Incinerator	Catalytic Incinerator
Operating time (hr/yr)	1,300	1,820	4,160	4,160
Capital Cost ($)	31,000	50,000	48,000	70,000
Annual Cost ($/yr)	13,500	12,900	55,200	27,100
Annual Cost per pound carbon removed ($/lb)	2.08	1.98	0.33	0.16

INDUSTRIAL SURFACE COATINGS

Industrial surface coatings refers to the application of protective or decorative coatings. Frequently, the coating is an organic solvent-based paint, which is applied by a spray gun in a paint spray booth. The freshly painted surface is then dried in an oven where the solvent which normally comprises about 60 percent of the paint volume, is evaporated. The size of paint spray operations ranges from as low as one gallon per day, to as high as several hundred gallons per day. According to the 1967 Department of Commerce census, industrial users consume about two-thirds of the paint manufactured in the United States.

Emissions

Insurance regulations require that paint spray booths be vented to the atmosphere to prevent the build-up of solvent vapor concentrations within the enclosure to more than 25 percent of the lower explosive limit.[2] Consequently, paint spray booths are normally ventilated at a rate of 100 to 150 fpm per square foot of booth opening.[2] Aerosols resulting from overspray in the spray booth, are removed by filters and water washers, to prevent their release to the atmosphere. However, these devices normally have little effect on solvent vapors. Although paint spray booths are a source of hydrocarbon emissions, the quantity of these emissions depends primarly on the degree of overspray within the booth, which can vary from as little as 5 to 10 percent, to as much as 80 to 90 percent.[2] Normally however, the degree of overspray in the paint spray booth is kept relatively low and the drying ovens are the major source of hydrocarbon emissions.

Control Systems and Costs

Incineration of the solvent vapors contained in the exhaust gases from

the paint spray booths and the drying ovens, is the control technique normally employed to limit emissions from industrial paint spraying operations. The costs of incineration are a funciton of the gas volume and small paint spraying operations would probably experience emission control costs similar to those shown for the graphic arts industry in the previous section. Many surface coating operations however, exhaust much larger volumes and the emission control costs would be greater. For example, the capital cost for an incinerator on both the paint spray booths and the drying ovens of a large paint spray line emitting about 45 pounds of solvent per hour, is estimated to be in the range of $400,000.[12] In general however, data to estimate annualized costs for both large and small paint spray operations are lacking.

Currently, within the domestic paint manufacturing industry, a great deal of effort is being devoted to the development of new paint formulations which require limited amounts of hydrocarbon solvents. While some of these new formulations can be used for certain coating applications, a number of limitations prevent them from being used for all applications. However, their use is rapidly increasing within the industry and if faced with the alternative of incineration, the use of these new formulations is expected to accelerate.

Water-based paints have been used for many years as architectural coatings and some manufacturers currently dip-coat and flow-coat parts with these paints. The use of water-based paints can result in a major decrease in hydrocarbon solvent emissions. If auto bodies are primed by electrodeposition for example, where the paint formula is about 90 percent solids and 10 percent water, emissions are reduced by about 90 percent over conventional spraying.[13]

Converting to the electrodeposition process however, requires completely scrapping existing paint spraying equipment and involves a large capital investment in new equipment. Elaborate washing systems and surface preparation systems are required, in addition to the basic electrodeposition equipment. Color change is impractical during assembly-line operation and the coated surface normally has a dull appearance which does not meet the gloss standards necessary on many products. Consequently, electrodeposition can be used only for primer coats in most applications.

Although finish coats of high gloss appearance can not be applied by electrodeposition, the application of water-based paints for finished coatings by spraying shows promise. The General Motors assembly plant in Los Angeles is converting to a water-based paint spray system. Complete conversion is expected by August 1974.[13] Since water evaporation rate is critical to the surface appearance, complex drying ovens and air conditioning equipment are being installed and as a result, power requirements may increase as much as 50 percent at this plant.[13]

In assessing the technical availability of water-based paint spray systems for applying finish coats of high gloss appearance, it must be kept in mind that this General Motors assembly plant in Los Angeles is the first major installation to attempt the use of water-based paints for finish coats. Consequently, until the plant has started up and resolved the problems that may arise, the use of water-based paint spray systems to apply finish coats must be considered as currently under development for most applications.

Powder coatings are applied by electrostatic spraying of fluidized bed techniques. The coated objects are then heated to about 450°F. This

melts the powder into a solid surface film. Most powder coatings consist of various plastics such as vinyls, acrylics, polyesters, epoxies, nylons and butrates. The application of coatings in this manner reduced hydrocarbon emissions by more than 90 percent, compared with conventional solvent-based coatings. One drawback however, is the high baking temperature. This prevents preliminary assembly of plastic or rubber parts prior to coating. While most powder coatings lack the weather resistence of conventional paints, they do excell on parts which are not exposed to the outdoors and where orange peel effects are not objectionable. Although powder coatings are finding use in some specific applications, water-based coating are generally considered to be more acceptable for wide-spread application than powder coatings at present. As the technology of powder coatings is developed however, they are expected to replace both water-based and conventional hydrocarbon solvent based coatings, as a result of lower investment costs due to a smaller oven and shorter process line, lower fuel costs for drying and baking, lower labor costs, and lower paint loss because overspray can be recycled.[12]

It is apparent, however, that the surface coatings area definitely needs a great deal of further study to determine the most feasible methods of control. Incineration can be viewed as the most straight forward method of control. The high investment costs for incineration equipment and the demand for large quantities of fuel however, make it an unattractive choice in many cases. In general, there is a serious lack of data available on investments, costs and time required to retrofit in this area, which make it difficult to develop compliance schedules and determine cost effectiveness.

PETROLEUM REFINERIES

General

There are literally hundreds of sources of hydrocarbon emissions in a modern petroleum refining complex, exclusive of the emissions from product storage and distribution. Prominent among these sources are waste water collection and disposal systems, blowdown systems, catalyst regenerators, compressors, pumps, valves, vacuum jets, asphalt air blowing, and process heaters and boilers. Although recent efforts have been directed toward identifying emission levels from a number of these sources, emissions will vary widely from refinery to refinery. In addition, wide variations are possible within a specific refinery depending on the mechanical condition of critical items of equipment, as well as differences in operating conditions.

The emission control systems that could be used to reduce emissions from many of these sources are well demonstrated and should be an integral part of any new refinery design and construction. Retrofit of many of these control systems in existing refineries with a few exceptions, should be relatively simple and require minimal lead times. However, with the exception of carbon monoxide boilers, which are primarily used to reduce carbon monoxide emissions from fluid catalytic cracking unit catalyst regenerators, the data is not available to estimate installation costs and control effectiveness of these systems.

Plans for new refinery construction merit careful review and existing refineries merit careful inspection to determine the adequacy of emission control equipment and in particular, the maintenance practices of the operator. For example, the collection and disposal of volatile hydrocarbons

emitted from waste-water collection and disposal equipment, can be accomplished by sealing the sewers and junction boxes and venting the vapors to an incinerator. Hydrocarbon emissions can be reduced from centrifugal pumps by the use of mechanical seals rather than soft packing (rubber, leather, etc.) to prevent leakage. Hydrocarbon emissions from vacuum jets, which are commonly used on crude oil vacuum distillation towers, can be collected and incinerated in process heaters and boilers, or sent to sulfur recovery units if the sulfur dioxide or hydrogen sulfide content of the non-condensibles is appreciable. These few examples illustrate the need for careful review and inspection.

Petroleum Product Liquid Storage

Evaporative emissions of hydrocarbons from storage tanks occur in all phases of the petroleum industry, from production of crude oil through final storage of finished products in the distribution and marketing system. As a result of legislative enactments and economic incentives in some cases, the industry has achieved a reasonable degree of control over hydrocarbons emissions from storage tanks. Most studies report that conservation measures are currently practiced on about 75 percent of the liquid hydrocarbon storage capacity in the petroleum industry. Nevertheless, it has been estimated that national emissions from petroleum product storage could be reduced by 50 percent if present control practices were extended to the remaining uncontrolled tankage.

Emissions

Hydrocarbon emissions from storage tanks depend essentially on three basic mechanisms: breathing loss, working loss, and standing storage loss. Breathing and working losses are associated with cone or fixed roof tanks, variable vapor space tanks, and low pressure tanks. Standing storage losses are generally associated with floating roof tanks. The magnitude of emissions depends on factors such as the physical properties of the material being stored, climatic conditions, and the size, type, control, and condition of the storage tank. An addendum to the second edition of the EPA publication "Compilation of Air Pollutant Emission Factors", (AP-42, April 1973), is being published which will provide guidance to local control agencies in estimating hydrocarbon emissions from various types of tankage storing numerous products under varying conditions.

Control Systems and Costs

The emission control system commonly employed within the industry to meet various State and local regulations which limit emissions from hydrocarbon storage facilities, is the "floating roof" tank. This tank is generally acceptable up to hydrocarbon vapor pressures under actual storage conditions, of 11.0 - 12.0 psia. Beyond this range, more effective measures are necessary. These normally consist of pressure tanks equipped with emergency relief valves, which prevent emissions to the atmosphere under normal conditions, or vapor recovery systems. Vapor recovery systems may afford a somewhat higher degree of control than the floating roof, if compressors are spared. Capital and operating costs for vapor recovery systems are normally considerably higher than either pressure tanks or "floating roof" tanks. However, the necessary data to make reasonable cost comparisons are lacking.

Due to wide-spread adoption of the "floating roof" tank within the industry, investment costs and costs effectiveness for this type of liquid hydrocarbon storage will be considered. Investment costs are presented in Table 6.

Table 6

Investments - Floating Roof vs. Fixed Roof Tanks[14]

Tank Size		Cost to Retrofit Fixed Roof to:		Δ Cost for New Construction Floating roof type vs. Fixed roof		
Diameter in feet	Normal Capacity bbls[a]	Covered Floating Roof	Internal Floating Cover	External Pontoon Floating Roof	Covered Floating Roof	Internal Floating Cover
20	1,100	--	$2,800	--	--	$ 2,800
40	9,000	$17,000	$6,500	--	n.a.	$ 7,000
60	22,000	$26,000	$12,000	+ $20,000	+ $17,000	+$12,000
90	54,000	$42,000	$22,000	+ $25,000	+ $23,000	+$24,000
110	80,000	$53,000	$40,000	+ $27,000	+ $33,000	--

[a] 42 gals/bbl

Construction considerations usually dictate that internal floating covers be used for small size tanks and covered floating roofs for intermediate size tanks. Extremely large tanks usually use the external type of floating roof, either double-deck or single-deck pontoon.

The emission control costs associated with storing a high volatility material such as gasoline and a low volatility material such as jet naphtha, in floating-roof tanks rather than fixed-roof tanks, are shown in Table 7.

Table 7

Control Costs for Retrofitting Fixed Roof Tanks[14] to Covered Floating Roofs

Product	Gasoline			Jet Naphtha		
Tank Size (10^3 bbls)	1.1	22	80	1.1	50	80
Investment Floating vs. Fixed (10^3\$)	2.8	26.0	53.0	2.8	26.0	53.0
Annualized Cost[a] (10^3\$)	(0.2)	(1.9)	(15.0)	0.3	2.4	2.8
Cost per gallon thruput-¢/gal	(0.03)	(0.03)	(0.03)	0.05	0.02	0.01

[a]Savings represented by parentheses.

The storage of gasoline in floating-roof tanks rather than fixed-roof tanks, results in a slight savings, while the storage of the less volatile jet naphtha in floating-roof tanks rather than fixed roof tanks, results in a slight cost.

Gasoline Distribution

A typical gasoline distribution pattern consists of delivery by pipeline, barge, or tanker to intermediate storage, then by pipeline or barge to bulk terminals. From the bulk terminal, the gasoline is delivered by tank truck or trailer to the service station or to a bulk plant and then by tank truck or trailer to the service station. At the service station, the gasoline is transferred from the tank trucks into underground storage tanks which are vented to the atmosphere. Automobiles are refueled from the underground tanks via the service station pump island.

Emissions

In uncontrolled bulk terminal tank truck loading operations, service station deliveries and automobile refueling, the vapors displaced by the gasoline are normally released directly to the atmosphere. The amount of hydrocarbons which are contained in these vapors is highly variable depending on the manner in which the tank truck is loaded at the terminal; the amount of gasoline contained in the vapors carried by the returning truck; the manner in which the gasoline is delivered to the service station; operator practice in automobile refueling; and a number of other factors such as physical properties of the gasoline; geographical and seasonal effects; and meteorological conditions.

To estimate the emissions and control costs associated with reducing hydrocarbon emissions from gasoline distribution, a hypothetical gasoline marketing system has been examined. This consists of: 300 gasoline service stations each dispensing 250,000 gallons per day and 15 tank trucks equipped with top loading to service the service stations. Emissions from this

distribution and marketing system under normal summer operating conditions are shown in Table 8.

Table 8

Hydrocarbon Emissions from an Uncontrolled Gasoline Distribution System[a]

	300 Service Stations[15] 25,000 gal/month per station		Bulk Terminal[16] 250,000 gal/day	
	10^3 lbs/yr	gals per 10^3 gals handled	10^3 lbs/yr	gals per 10^3 gals loaded
Loading loss	---	---	520	1.10[b]
Tank Truck Delivery	671	1.44	--	--
Breathing loss	77	0.17	--	--
Diurnal Loss	neg.	--	--	--
Subtotal	748	1.61	520	1.10
Automobile Refueling	675	1.44	--	--
Total	1,423	3.05	520	1.10

[a]Excludes tankage evaporative losses from bulk plant and terminal

[b]Submerged loading.

Control Systems and Costs

Achieving a substantial reduction in emissions throughout the gasoline distribution system requires emission controls at gasoline service stations. This in turn requires vapor recovery during tank truck loading at bulk plants and terminals. The most "feasible" approach for reducing emissions by 85 to 90 percent is as follows:

Phase I (55 percent control-short range):

1. Modify tank trucks and install the necessary equipment at service stations to recycle the vapors displaced from the underground storage tanks at service stations back to the tank truck during gasoline delivery.
2. Install vapor recovery systems to collect and recover the vapors collected at the service station, as they are displaced from the tank truck during reloading at bulk plants and terminals.

Phase II (30 to 35 percent control-long range):

1. Install special gasoline dispensing nozzles, hoses and vapor return lines on the gasoline pumps at the service stations to collect and return vapors displaced from the automobile fuel tank during refueling, to the underground storage tanks at the service station.
2. Install vapor recovery equipment on the underground storage tanks at the service station if necessary, to achieve an overall reduction in emissions of 90 percent at the service station.

Capital investments, annual costs, and cost effectiveness, on a per station basis are summarized in Table 9. These estimates are based on retrofit, since the number of existing service stations and loading racks far outnumber anticipated new construction.

Table 9

Control Costs per Service Station Dispensing[15,17,18,19]
25,000 Gal/month of Gasoline

Phase I (55 Percent Reduction in Emissions)

	Recycle Vapor To Tank Truck	Vapor Recovery at Bulk Terminal	Phase I Total
Investment($)	1,073	783	1,856
Emission Reduction			
(lbs/yr)	1,933	1,733	3,666
(gal/yr)	372	334	706
Annual Cost			
($/yr)	97	70	167
($/lb recovered)	0.05	0.04	0.045
(¢/gal pumped)	0.03	0.02	0.055

Phase II (35 Percent Reduction in Emissions)

	Carbon Canister	Refrigeration-Condensation
Investment ($)	6,090	12,678
Emission Reduction		
(lbs/yr)	2,300	2,300
(gal/yr)	443	443
Annual Cost		
($/yr)	1,298	2,693
($/lb recovered)	0.56	1.17
(¢/gal pumped)	0.43	0.89

Table 9 (cont.)

Phase I + II (90 Percent Reduction in Emissions

	Carbon Canister	Refrigeration-Condensation
Investment ($)	7,946	14,534
Emission Reduction		
(lbs/yr)	5,966	5,966
(gal/yr)	1,149	1,149
Annual Cost		
($/yr)	1,465	2,860
($/lb recovered)	0.24	0.48
(¢/gal pumped)	0.48	0.95

The quality of the Phase I investment data and emission control efficiencies are considered "moderate". Recycle of the vapors displaced from the underground storage tanks at the service station, to the tank truck during gasoline delivery, is feasible and has been demonstrated. Vapor recovery systems to collect and recover the vapors displaced from the tank trucks at bulk terminals during reloading, have proven feasible during warm weather operations. However, operability and safety in cold weather conditions are still in question. Nevertheless, Phase I could be implemented at this time.

The quality of the Phase II data should be considered "questionable". Investments would be much lower if a simple displacement vapor collection and recycle system could be made to work successfully during automotive refueling, without requiring the installation of vapor recovery facilities. The use of carbon canisters or refrigeration-condensation techniques, are two alternative approaches for vapor recovery and vendors of this equipment are limited. Special vapor collection and return nozzles, to effect a tight-fit between the gasoline dispensing nozzle and the automobile fuel tank fill pipe, are required and these are still in the development stage. With many existing vehicles and available nozzle designs a tight-fit is not possible because of location or design of the automobile fuel tank fill pipe. Vacuum-assist, which could be used to overcome some of the disadvantages of a lack of tight-fit between the nozzle and the automobile fill pipe, has been identified by the industry as a possible explosive hazard.

PETROCHEMICALS

Carbon Black

Most of the carbon black produced in the United States is produced by the furnace process (83 percent).[20] Although the thermal process and the channel process are also used, they account for only a small amount of the total carbon black production (14 percent and 3 percent, respectively).[20] The thermal process however, is not a major source of emissions and the channel process is viewed as obsolete within the industry. Consequently, the furnace process is of most concern in terms of pollutant emissions.

In this process, natural gas and heavy oil are introduced into a furnace with a limited amount of air. A combination of cracking and combustion occurs, which leads to the production of carbon. The hot gases from the furnace are cooled and passed through bag filters in which the carbon is collected. The gases are then normally discharged directly to the atmosphere, although in some cases, the waste gases are combusted before release to the atmosphere.

Emissions

The production of carbon black by the furnace process can be a major source of emissions. It has been estimated for example, that about 14,000 pounds per hour of carbon monoxide and 700 pounds per hour of various hydrocarbons could be emitted to the atmosphere from an uncontrolled carbon black plant producing 90 million pounds of carbon black a year, using the furnace process.[20]

Control Systems and Costs

Waste heat boilers, thermal incinerators or catalytic incinerators could be used to reduce emissions by about 100 percent.[20] Plume burners

which are a type of flare, are somewhat less efficient, although they could be used to reduce emissions by about 90 percent.[20] Only one carbon black plant in the United States uses an incinerator waste heat boiler, although several are equipped with plume burners.[20]

The most effective emission control technique for a new plant would be the use of a thermal incinerator or a waste heat boiler. No supplemental or auxillary fuel would be required in either case. Heat recovered in a waste heat boiler could be used to preheat the incoming gas and to generate the steam necessary to operate the plant. Although flame control could be difficult in some cases, flameouts could be prevented if adequate burner controls were provided. For a new 90 million lbs/yr carbon black plant using the furnace process, the installation of a waste-heat boiler would increase the capital investment by about $720,000.[20] However, the waste-heat boiler would have a net operating savings of $10,500 per year, due to the credit for steam generation.[20]

A plume burner would actually be more costly and less effective. Although the initial capital investment would only be increased by about $150,000, the net capital costs would be about $33,000 per year.[20] This is equivalent to an increase of 0.04¢/lb in production costs.[20]

Acrylonitrile

The use of the Sohio propylene ammoxidation process is universal within the United States for the production of acrylonitrile.[21] Vaporized propylene and ammonia are mixed with air and steam and introduced into a catalytic reactor. The gaseous reaction products are withdrawn and processed through a countercurrent absorber where the acrylonitrile is absorbed in water. The gases are then vented directly to the atmosphere in many cases. The aqueous acrylonitrile solution is fractionated to recover the acrylonitrile.

Emissions

The production of acrylonitrile can be a major source of emissions of carbon monoxide and various hydrocarbons. Table 10 summarizes the estimated emissions released to the atmosphere from an uncontrolled acrylonitrile plant producing 200 million pounds of acrylonitrile per year.

Table 10

Estimated Emissions from a 200 Million Pounds per Year Acrylonitrile Plant[21]

Carbon Monoxide	3,000 lbs/hr
Propane and Propylene	1,400 lbs/hr
Hydrogen Cyanide	10 lbs/hr
Acrylonitrile	10 lbs/hr
Acetonitrile	160 lbs/hr

Control Systems and Costs

Waste heat boilers, thermal incinerators or catalytic incinerators could be used to reduce hydrocarbon emissions by about 100 percent.[21]

Plume burners are generally considered to be less efficient, although it is estimated that they could reduce emissions by about 90 percent.[21] None of the acrylonitrile plants in the United States currently operate any control equipment to limit emissions.[21] However, waste-heat boilers, thermal incinerators, catalytic incinerators and plume burners are in use on similar processes and could be installed at acrylonitrile plants.

A large amount of steam is produced in most acrylonitrile plants, consequently, most of the steam that could be produced in an incinerator-waste heat boiler used to reduce emissions, would have to be exported. The heating value of the pollutant concentrations in the waste gases is normally quite low and a supplemental or auxillary fuel would have to be used to achieve complete combustion. This method of emission control for a typical 200 million lbs/yr acrylonitrile plant would cost about $115,000, assuming a steam credit of 75¢/thousand pounds.[21]

Probably the most feasible emission control device is a thermal incinerator. Although a small amount of supplemental fuel would be required, heat could be recovered to preheat the propylene, ammonia and combustion air introduced into the acrylonitrile reactor. An incinerator for a typical 200 million lbs/yr acrylonitrile plant could cost about $350,000 initially and would have a total net annual operating cost of about $135,000.[21]

Formaldehyde

All the formaldehyde produced in the United States is manufactured by two methods, both of which use methanol as the raw material.[22] The first uses an iron-oxide catalyst and a large excess of air. The second uses a silver catalyst and requires only about one-eighth as much air as the iron-oxide process. In both methods, the formaldehyde is produced in a reactor and then absorbed from the exit gases by a water scrubber. The scrubber off-gases are normally vented directly to the atmosphere. The formaldehyde is marketed as a water solution, although it is purified before sale.

About 22 percent of the domestic formaldehyde production is based on the iron-oxide process and about 78 percent based on the silver process.[22]

Emissions

The release of the absorber off-gases directly to the atmosphere is the primary source of emissions associated with the production of formaldehyde. In a plant using an iron-oxide catalyst, the composition of these off-gases may vary considerably, depending on the degree to which they are recycled within the plant. Most of the iron-oxide plants currently operating however, operate with maximum recycle.[22] In a plant using a silver catalyst, the recycle of absorber off-gases is not general practice, since it offers no advantages to the operator as it does in a plant using an iron-oxide catalyst.

Estimated emission rates from an uncontrolled formaldehyde plant producing 100 million lbs/yr of formaldehyde as a 37 percent aqueous solution, are presented in Table 11. The emissions from the plant using an iron-oxide catalyst are based on maximum recycle of the absorber off-gases.

Table 11

Emissions from an Uncontrolled 100 Million lbs/yr Formaldehyde Plant[22]

	Iron-oxide Catalyst	Silver Catalyst
Formaldehyde (lbs/hr)	5	10
Methanol (lbs/hr)	20	30
Dimethyl Ether	10	--
Hydrogen (lbs/yr)	--	120
Carbon Monoxide (lbs/hr)	--	60

Control Systems and Costs

Thermal incineration of the absorber off-gases is the most feasible method of reducing emissions from formaldehyde plants which use an iron-oxide catalyst. Although some auxillary fuel is required to ensure complete combustion, there is normally not enough heat released to make steam generation attractive. A plume burner could be used rather than a thermal incinerator, but would be somewhat less efficient in reducing emissions. A water scrubber could also be used to remove about 95 percent of the formaldehyde and methanol from the absorber off-gases, however, none of the dimethyl ether would be removed.[22]

One iron-oxide formaldehyde plant currently operating in the United States, employs a water scrubber to reduce emissions from the absorber. None however, use thermal incinerators, although this is not due to any technical problems which might prevent their use. The use of an incinerator at a 100 million lb/yr iron-oxide formaldehyde plant would cost about $44,000 initially and the operating cost would be about $35,000 per year.[22]

The most feasible method of reducing emissions from formaldehyde plants which employ a silver catalyst, would be to combust the absorber off-gases

in the utility boilers associated with the formaldehyde plant, or chemical complex. The absorber off-gases from the silver process have a much higher heating value than those from the iron-oxide process and could be combusted without auxilliary fuel. However, because of the relatively small volume of gases the installation of a separate waste-heat boiler is not practical. This approach would cost about $45,000 initially, to make the necessary modifications to the utility boilers, but would show a net operating savings of about $3,000 per year.[22]

The use of a plume burner would be less costly initially, requiring an outlay of only about $20,000.[22] However, the net operating cost would be about $5,500 per year.[22]

Ethylene Dichloride

In 1970 about 55 percent of all ethylene dichloride was produced by oxychlorination of ethylene.[23] The remaining 45 percent was produced by chlorination of ethylene.[23] However, the ethylene chlorination process is relatively unpolluting compared to the oxychlorination process and therefore is not of major interest here.

In the oxychlorination process, ethylene, air and hydrochloric acid are introduced into a reactor. In the presence of a catalyst, these materials react to yield ethylene dichloride. The ethylene dichloride is absorbed from the reactor effluent gases and the gases are normally vented directly to the atmosphere.

There are a number of variations in the basic oxychlorination process. Although one plant uses oxygen rather than air in the reactor, the reaction sections are similar. The ethylene dichloride recovery systems, however, vary significantly from plant to plant and thus emissions to the atmosphere vary from plant to plant.

Emissions

Estimated emissions from a typical uncontrolled oxychlorination plant producing 700 million lbs/yr of ethylene dichloride are summarized in Table 12.

Table 12

Emissions from an Uncontrolled 700 Million lb/yr Ethylene Dichloride Plant[23]

Carbon Monoxide	600 lbs/hr
Methane	500 lbs/yr
Ethylene	400 lbs/hr
Ethane	600 lbs/hr
Ethylene Dichloride	600 lbs/hr
Ethylene Chloride	500 lbs/hr

Control Systems and Costs

Incineration with or without steam generation or heat recovery, followed by a water or dilute caustic scrubber would reduce hydrocarbon (and chlorine) emissions by about 100 percent.[23] However, none of the oxychlorination plants currently operating, use incineration to reduce emissions. Heat recovery or steam generation from incineration would be difficult because of the corrosive nature of the hydrochloric acid formed.

The installation of a waste heat boiler and caustic scrubber in a typical 700 million lbs/yr plant would cost about $900,000 initially, and the net operating costs would be about $300,000 per year.[23] The use of a thermal incinerator and scrubber would only cost about $63,000 initially, however, the net operating cost would be about $400,000 per year.[23] It should be noted that due to the corrosive environment existing in the waste heat boiler or incinerator, special alloy materials might be required and these estimated costs could be in error by a considerable margin.

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